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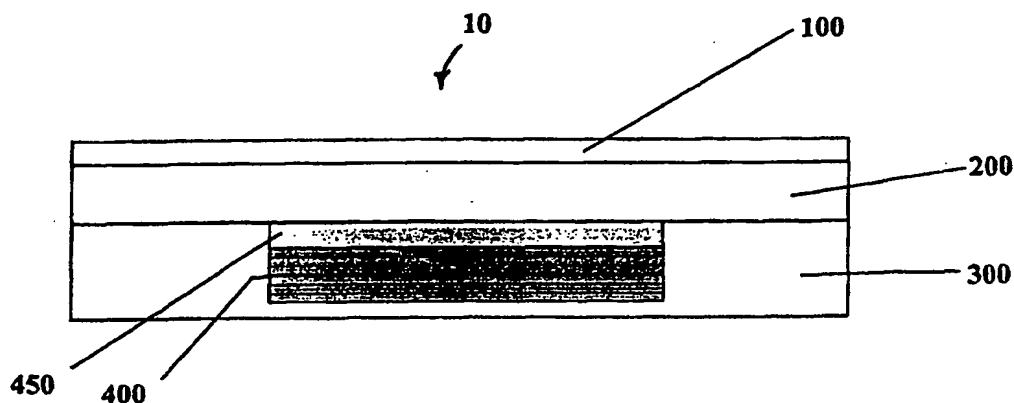
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(21) International Application Number: PCT/US99/20650 (22) International Filing Date: 10 September 1999 (10.09.99) (30) Priority Data: 60/099,939 11 September 1998 (11.09.98) US (71) Applicant (for all designated States except US): FED CORPORATION [US/US]; Hudson Valley Research Park, 1580 Route 52, Hopewell Junction, NY 12533 (US). (72) Inventor; and (75) Inventor/Applicant (for US only): PICHLER, Karl [AT/US]; 18 Hillside Avenue, Wappingers Falls, NY 12590 (US). (74) Agent: COYNE, Patrick, J.; Collier Shannon Rill & Scott, PLLC, Suite 400, 3050 K Street, N.W., Washington, DC 20007 (US).		(81) Designated States: US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>

(54) Title: TOP EMITTING OLED WITH REFRACTORY METAL COMPOUNDS AS BOTTOM CATHODE



(57) Abstract

A top emitting organic light emitting device (10) includes a substrate (300), a cathode conductor layer (400) having a low work function refractory metal layer (450), an organic light emitting material layer (200), and an anode layer (100). The low work function refractory metal layer (450) preferably is selected from a nitride, boride, or carbide of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.

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TOP EMITTING OLED WITH REFRACTORY METAL COMPOUNDS AS BOTTOM CATHODE

Cross Reference To Related Patent Application

This application relates to and claims priority on United States provisional application serial number 60/099,939, filed September 11, 1998.

Field of the Invention

The present invention relates to an Organic Light Emitting Device ("OLED") video display structure for a color video display or high brightness monochrome display and methods of making such. More specifically, the present invention relates to a top emitting OLED with a bottom cathode.

Background of the Invention

Organic light emitting devices have been known for approximately two decades. OLEDs work on certain general principles. An OLED is typically a laminate formed on a substrate such as soda-lime glass or silicon. A light-emitting layer of a luminescent organic solid, as well as adjacent semiconductor layers, are sandwiched between a cathode and an anode. The semiconductor layers may be hole-injecting or electron-injecting layers. The light-emitting layer may be selected from any of a multitude of fluorescent organic solids. The light-emitting layer may consist of multiple sublayers or a single blended layer.

When a potential difference is applied across the device, negatively charged electrons move from the cathode to the electron-injecting layer and finally into the layer(s) of organic material. At the same time positive charges, typically referred to as holes, move from the anode to the hole-injecting layer and finally into the same organic light-emitting layer(s). When the positive and negative charges meet in the layer(s) of organic material, they produce photons.

The wave length -- and consequently the color -- of the photons depends on the material properties of the organic material in which the photons are generated. The color of

light emitted from the OLED can be controlled by the selection of the organic material, or by the selection of dopants, or by other techniques known in the art. Different colored light may be generated by mixing the emitted light from different OLEDs. For example, white light is produced by mixing blue, red, and green light simultaneously.

5 Either the OLED anode or the cathode (or both) should be transparent in order to allow the emitted light to pass through to the viewer. The cathode may be constructed of a low work function material and the holes may be injected from the anode, a high work function material, into the organic material via a hole transport layer.

10 Typically, OLEDs operate with a DC bias in the range of 2 to 30 volts. The OLED brightness may be controlled by adjusting the voltage or current supplied to the anode and cathode. The relative amount of light generated is commonly referred to as the "gray level." OLEDs may work better when operated in a current mode. The light output may be more stable for constant current drive than for a constant voltage drive. This is in contrast to many other display technologies, which are normally operated in a voltage mode. As a result, an
15 active matrix display using OLED technology requires a specific pixel architecture to provide for a current mode of operation.

 In a matrix-addressed OLED device, numerous individual OLEDs may be formed on a single substrate and arranged in groups in a grid pattern. Several OLED groups forming a column of the grid may share a common cathode, or cathode line. Several OLED groups
20 forming a row of the grid may share a common anode, or anode line. The individual OLEDs in a given group emit light when their cathode line and anode line are activated at the same time. A group of OLEDs within the matrix may form one pixel in a display, with each OLED usually serving as one subpixel or pixel cell.

 OLEDs have a number of beneficial characteristics. These include: a low activation
25 voltage (about 5 volts); fast response when formed with a thin light-emitting layer; high brightness in proportion to the injected electric current; high visibility due to self-emission; superior impact resistance; and ease of handling of the solid state devices in which they are used. OLEDs, have practical application in television, graphic display systems, and digital printing. Although substantial progress has been made in the development of OLEDs

to date, additional challenges remain. For example, OLEDs continue to face a general series of problems associated with their long-term stability. In particular, there is a need to provide a stable low work function cathode that is both a good electron injector for OLEDs and compatible with standard semiconductor processing. It would be particularly beneficial to provide an OLED cathode structure that is stable in an air ambient, so that semiconductor processing methods that are carried out in air may be utilized. Low work function cathodes currently in use with OLEDs have not demonstrated the required stability.

There is also a need for a cathode manufacturing process that can be integrated with standard silicon chip manufacturing processes. Due to resolution requirements it is not feasible to deposit the cathode onto the substrate after substrate manufacturing is complete (e.g., through a shadow mask process and as a first step of the deposition processes for the OLED stack). This has been a problem due to the very high resolution requirements of certain flat panel displays (approximately less than 20 micrometer).

The present invention meets the needs set forth above and provides other benefits as well.

Objects of the Invention

Therefore it is an object of the present invention to provide a stable top emitting OLED with a bottom cathode.

It is another object of the present invention to provide an economical and uncomplicated manufacturing process for an OLED.

It is a further object of the present invention to provide a stable low function cathode directly overlying the silicon substrate or wafer of an OLED.

It is still a further object of the present invention to provide a process for integrating the construction of a stable low work function cathode into the semiconductor processing methods used to make OLEDs.

It is yet a further object of the present invention to provide an OLED with a stable cathode layer capable of being manufactured and packaged in conjunction with a silicon wafer or substrate and stored, shipped, handled, and processed in standard conditions, such as an air ambient before deposition of the organic layer(s).

Additional objects and advantages of the invention are set forth, in part, in the description which follows and, in part, will be apparent to one of ordinary skill in the art from the description and/or from the practice of the invention.

Summary of the Invention

5 In response to this challenge, Applicant has developed an innovative, economical OLED comprising: a substrate; a low work function refractory metal layer overlying said substrate; a stack of one or more layers of light emitting organic material overlying said low work function refractory metal layer; and an anode conductor layer overlying said organic material.

10 Applicant has further developed an OLED comprising: a substrate; a cathode conductor layer overlying said substrate; a low work function refractory metal layer overlying said substrate; a stack of one or more layers of light emitting organic material overlying said low work function refractory layer; and an anode conductor layer overlying said organic material.

15 Applicant has still further developed an OLED comprising: a substrate; a cathode conductor layer overlying said substrate; a low work function refractory metal layer overlying said substrate; an insulator layer overlying said low work function refractory metal layer; a stack of one or more layers of light emitting organic material overlying said insulator layer; and an anode conductor layer overlying said organic material.

20 Applicant has also developed a method of making an OLED comprising the steps of: providing a substrate; forming a low work function refractory metal layer overlying said substrate; forming a stack of one or more organic materials overlying said low work function refractory metal layer; and forming an anode layer overlying said organic materials.

25 Applicant has also developed a method of making an OLED comprising the steps of: providing a substrate; forming a cathode conductor layer overlying said substrate; forming a low work function refractory metal layer overlying said cathode conductor layer; forming a stack of one or more organic materials overlying said low work function refractory metal layer; and forming an anode layer overlying said organic materials.

Applicant has also developed a method of making an OLED comprising the steps of: providing a substrate; forming a cathode conductor layer overlying said substrate; forming a low work function refractory metal layer overlying said cathode conductor layer; forming an insulator layer overlying said low work function refractory metal layer; forming a stack of one or more organic materials overlying said insulator layer; and forming an anode layer overlying said organic materials.

Applicant has also developed a method of making a substrate and cathode structure adapted to be handled in an air ambient and for use in a top emitting OLED having a top anode, the method comprising the steps of: providing a planar substrate material selected from the group consisting of silicon and glass; forming a cathode conductor layer overlying said substrate; and forming a low work function refractory metal layer overlying said cathode conductor layer.

Applicant has also developed a method of making a substrate and cathode structure adapted to be handled in an air ambient and for use in a top emitting OLED having a top anode, the method comprising the steps of: providing a planar substrate material selected from the group consisting of silicon and glass; and forming a low work function refractory metal layer overlying said substrate.

It is to be understood that both the foregoing general description and the following detailed description are exemplary and explanatory only, and are not restrictive of the invention as claimed. The accompanying drawings, which are incorporated herein by reference, and which constitute a part of this specification, illustrate certain embodiments of the invention, and together with the detailed description serve to explain the principles of the present invention.

Brief Description of the Drawings

Fig. 1 is a cross-section in elevation of an OLED according to a first embodiment of the present invention.

Fig. 2 is a cross-section in elevation of an OLED according to a second embodiment of the present invention.

Fig. 3 is a cross-section in elevation of an OLED according to a third embodiment of the present invention.

Fig. 4 is a cross-section in elevation of an OLED according to a fourth embodiment of the present invention.

5

Detailed Description of the Invention

Reference will now be made in detail to a first embodiment of the present invention, an example of which is illustrated in the accompanying drawings. The first embodiment of the present invention is illustrated by the OLED 10 shown in Fig. 1. The OLED 10 of the first embodiment of the present invention may include a substrate 300, a top anode layer (transparent or semi-transparent) 100, a stack of one or more light emitting organic materials 200, a cathode conductor layer 400, and a low work function refractory metal layer 450.

Substrate 300 may be glass or part of a Si wafer which includes transistors and a plurality of pixels. The cathode conductor layer 400 may be a reflective metal or alloy, such as Ag, Al, or Cr. The reflective conductor layer 400 may enhance the brightness of the OLED by reflecting light generated in the organic material 200 through the anode layer 100 towards a viewer.

With reference to the embodiment shown in Fig. 1, the innovation provides a novel easy-to process (i.e., integration on Si-chip) and stable low work function bottom cathode. A range of refractory metal compounds may be used (such as nitrides, borides and/or carbides of, for example, Ti, Zr, Hf, Nb, W, Mo, Cr, Ta or V) in the refractory metal layer 450. The OLED 10 may include in particular a Si-chip substrate 300 with a built-in (pre-fabricated) pixelated TiN low work function refractory metal layer 450 as the OLED cathode.

The desired work function for the refractory metal layer 450 is below about 4 eV. It is preferable that the work function be below about 3.5 eV, and in some cases below about 3 eV. It is also preferable that the refractory metal compound selected be very stable in an air ambient. A study of these refractory metal materials is provided in "Field Emission Cold Cathode Devices Based on Eutactic Systems," by V.G. Rivlin and D. Stewart, European Office of Aerospace Research and Development, London, England and Fulmar Research

Institute, Stoke Poges, Slough SL2 4QD, England (prepared for U.S. Airforce Scientific Research, Bolling AFB, D.C. 20332 under contract/grant number: AFOSR-77-3292), incorporated by reference herein.

With continued reference to the embodiment shown in Fig. 1, the preferred low work function material for the bottom cathode is TiN. The use of TiN results in a stable low work-function bottom cathode that is easy to construct using well established semiconductor processes such as sputtering or chemical vapor deposition. TiN has a work function typically in the range of 2.5 to 3 eV. TiN may also provide a conductive plug in active matrix displays underlying an ITO (Indium Tin Oxide) layer to provide low resistance and highly stable (i.e. no oxidation) interconnects between the underlying electrodes and the ITO. TiN is a good conductor with a resistivity of approximately 21.7 microhm-cm.

The construction of the TiN layer 450 may be integrated into the Si-chip manufacturing process. The TiN layer 450 is environmentally stable allowing the Si substrate and the TiN layer to be handled in air, stored, shipped, cleaned, etc., without damage. Other low work function cathodes cannot be integrated with the Si substrate and then handled because the materials are not typically stable.

With reference to Fig. 2, in which like reference numerals refer to like elements in other figures, in an alternative embodiment of the invention the cathode conductor layer 400 may be eliminated if the low work function refractory metal layer 450 provides sufficient reflectivity to achieve the desired brightness for the OLED.

With reference to Fig. 3, in which like reference numerals refer to like elements in other figures, in another alternative embodiment of the invention the cathode conductor layer 400 and the low work function refractory metal layer 450 are provided on the planar upper surface of the substrate 300 instead of as a plug as shown in Fig. 1.

With reference to Fig. 4, in a further embodiment of the present invention in which like reference numerals refer to like elements shown in the other figures, an insulator layer of LiF, SiO, SiO₂, or some other dielectric oxide, nitride, or oxi-nitride may be provided overlying the refractory metal layer 450 and below the organic material 200.

With continued reference to Fig. 4, in some cases the OLED 10 may function better with a thin insulator layer 500 between the refractory metal layer 450 and the organics 200. The thin insulator layer may be deposited in conjunction with the construction of the refractory metal layer 450 and the substrate 300.

Presently, top-emitting OLEDs generally have been forced to employ a bottom-anode and top-cathode structure primarily due to the lack of stability of the cathode when on the bottom (i.e., on the Si wafer). This structure requires working with the top reactive cathode during the OLED deposition process. The use of a top cathode structure complicates the manufacturing process and limits the choice of cathode materials. Standard OLEDs may include bottom transparent anodes on glass and use top-opaque cathodes. TiN would be extremely difficult to use in this case because the deposition conditions for the TiN are likely to cause extensive damage to the underlying organic layer(s). The present innovation allows for top emitting OLEDs with transparent top-anode structures.

It will be apparent to those skilled in the art that various modifications and variations can be made in the construction, configuration, and/or operation of the present invention without departing from the scope or spirit of the invention. For example, in the embodiments mentioned above, the thicknesses of the various layers of the OLED, as well as the deposition processes used, may be widely varied without departing from the intended scope of the invention. Thus, it is intended that the present invention cover the modifications and variations of the invention provided they come within the scope of the appended claims and their equivalents.

WHAT IS CLAIMED IS:

1. An OLED comprising:
a substrate;
a low work function refractory metal layer overlying said substrate;
a stack of one or more layers of light emitting organic material overlying said low
5 work function refractory metal layer; and
an anode conductor layer overlying said organic material.
2. The OLED of Claim 1 wherein the material for the low work function refractory metal layer is selected from the group consisting of: nitrides, borides, and carbides of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.
3. The OLED of Claim 1 further comprising an insulator layer between said low work function refractory metal layer and said organic material.
4. The OLED of Claim 3 wherein the material for the insulator layer is selected from the group consisting of: LiF, SiO, and SiO₂.
5. The OLED of Claim 4 wherein the material for the low work function refractory metal layer is selected from the group consisting of: nitrides, borides, and carbides of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.
6. The OLED of Claim 1 further comprising a cathode conductor layer between said substrate and said low work function refractory metal layer.
7. The OLED of Claim 6 wherein the material for the cathode conductor layer is selected from the group consisting of: a reflective metal, and a reflective alloy.

8. The OLED of Claim 6 wherein the cathode conductor layer comprises a material selected from the group consisting of: Al, Ag, and Cr.

9. An OLED comprising:

a substrate;

a cathode conductor layer overlying said substrate;

a low work function refractory metal layer overlying said substrate;

5 a stack of one or more layers of light emitting organic material overlying said low work function refractory layer; and

an anode conductor layer overlying said organic material.

10. The OLED of Claim 9 wherein the material for the low work function refractory metal layer is selected from the group consisting of: nitrides, borides, and carbides of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.

11. The OLED of Claim 10 wherein the cathode conductor layer comprises a material selected from the group consisting of: Al, Ag, and Cr.

12. An OLED comprising:

a substrate;

a cathode conductor layer overlying said substrate;

a low work function refractory metal layer overlying said substrate;

5 an insulator layer overlying said low work function refractory metal layer;

a stack of one or more layers of light emitting organic material overlying said insulator layer; and

an anode conductor layer overlying said organic material.

13. The OLED of Claim 12 wherein the material for the low work function refractory metal layer is selected from the group consisting of: nitrides, borides, and carbides of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.

14. The OLED of Claim 13 wherein the cathode conductor layer comprises a material selected from the group consisting of: Al, Ag, and Cr.

15. The OLED of Claim 14 wherein the material for the insulator layer is selected from the group consisting of: LiF, SiO, and SiO₂.

16. A method of making an OLED comprising the steps of:
providing a substrate;
forming a low work function refractory metal layer overlying said substrate;
forming a stack of one or more organic materials overlying said low work function
5 refractory metal layer; and
forming an anode layer overlying said organic materials.

17. The OLED of Claim 16 wherein the material for the low work function refractory metal layer is selected from the group consisting of: nitrides, borides, and carbides of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.

18. A method of making an OLED comprising the steps of:
providing a substrate;
forming a cathode conductor layer overlying said substrate;
forming a low work function refractory metal layer overlying said cathode conductor
5 layer;
forming a stack of one or more organic materials overlying said low work function refractory metal layer; and
forming an anode layer overlying said organic materials.

19. The OLED of Claim 18 wherein the material for the low work function refractory metal layer is selected from the group consisting of: nitrides, borides, and carbides of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.

20. The OLED of Claim 19 wherein the cathode conductor layer comprises a material selected from the group consisting of: Al, Ag, and Cr.

21. A method of making an OLED comprising the steps of:
providing a substrate;
forming a cathode conductor layer overlying said substrate;
forming a low work function refractory metal layer overlying said cathode conductor
5 layer;
forming an insulator layer overlying said low work function refractory metal layer;
forming a stack of one or more organic materials overlying said insulator layer; and
forming an anode layer overlying said organic materials.

22. The OLED of Claim 21 wherein the material for the low work function refractory metal layer is selected from the group consisting of: nitrides, borides, and carbides of Ti, Zr, Hf, Nb, W, Mo, Cr, Ta, and V.

23. The OLED of Claim 22 wherein the cathode conductor layer comprises a material selected from the group consisting of: Al, Ag, and Cr.

24. The OLED of Claim 23 wherein the material for the insulator layer is selected from the group consisting of: LiF, SiO, and SiO₂.

25. The method of Claim 23 wherein the material for the insulator layer is selected from the group consisting of: LiF, an oxide, a nitride, and an oxi-nitride.

26. A method of making a substrate and cathode structure adapted to be handled in an air ambient and for use in a top emitting OLED having a top anode, the method comprising the steps of:

- 5 providing a planar substrate material selected from the group consisting of silicon and glass;
- forming a cathode conductor layer overlying said substrate; and
- forming a low work function refractory metal layer overlying said cathode conductor layer.

27. A method of making a substrate and cathode structure adapted to be handled in an air ambient and for use in a top emitting OLED having a top anode, the method comprising the steps of:

- 5 providing a planar substrate material selected from the group consisting of silicon and glass; and
- forming a low work function refractory metal layer overlying said substrate.

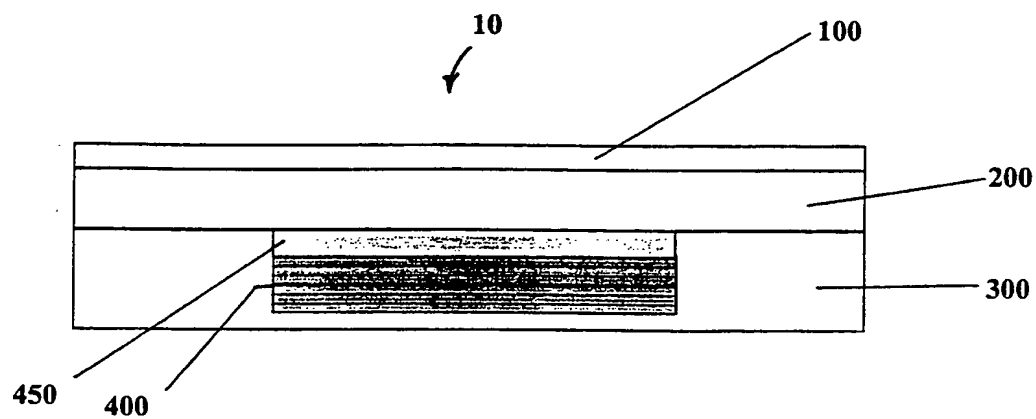


FIG. 1

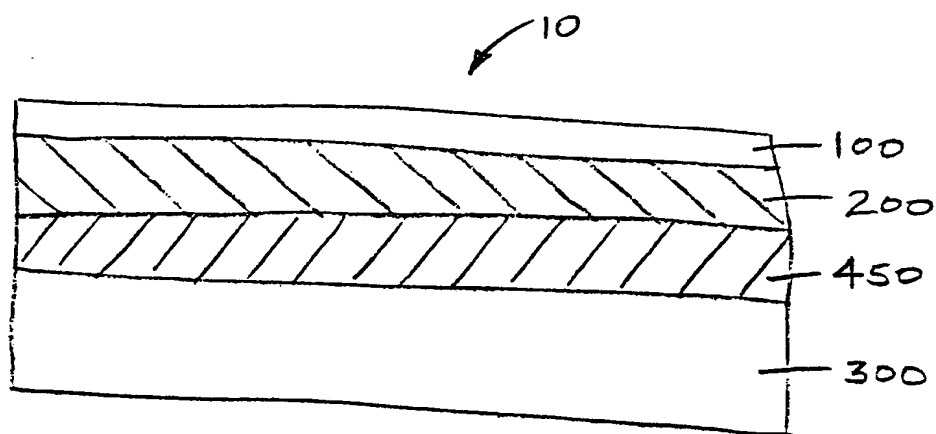


FIG. 2

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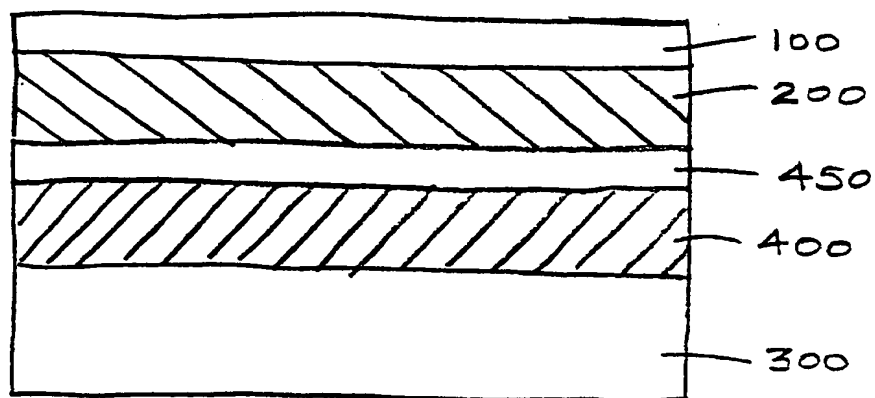


FIG. 3

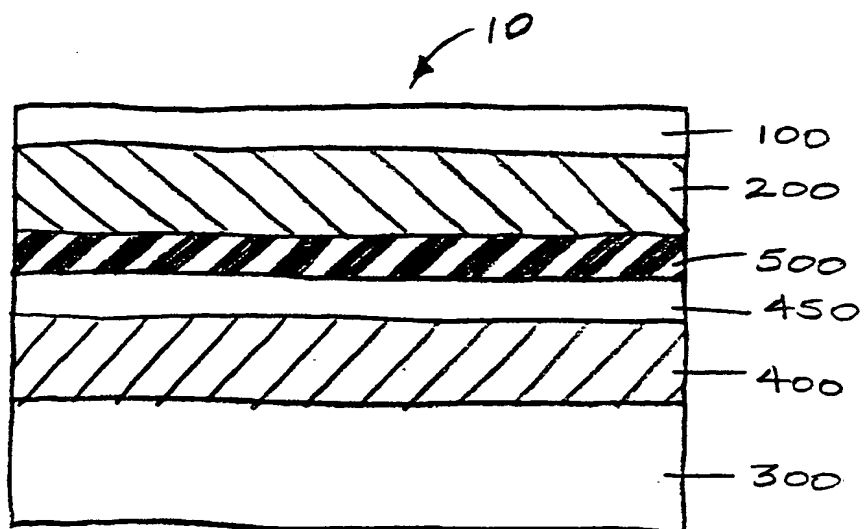


FIG. 4

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US99/20650

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :H01J 1/62

US CL :313/504

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 313/504, 503, 506, 509

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
NONE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X, E ----- Y, E	US 5,969,474 A (ARAI) 19 October 1999 (19.10.99) FIG. 1, col. 3, lines 44-50.	1, 2, 6-10, 16-20, 26, 27 ----- 3-5, 11-15, 21-25
X --- Y	JP 10125469 A (ARAI) 15 May 1998 (15.05.98) FIG. 1.	1, 2, 6-10, 16-20, 26, 27 ----- 3-5, 11-15, 21-25
A	US 5,164,799 A (UNO) 17 November 1992 (17.11.92) col. 2, lines 39-68.	1-27

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

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Date of the actual completion of the international search

24 NOVEMBER 1999

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US99/20650

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4,099,091 A (YAMAZOE et al.) 4 July 1978 (04.07.78) col. 2, lines 36-45.	3-5, 11-15, 21-25